Plasma-Chemical Oxygen-Iodine Laser: Problems of Development

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ABSTRACT

Great success has been obtained in the R&D of a chemical oxygen-iodine laser (COIL) operating on the electronic transition of the iodine atom, which gets an excitation from the energy donor -singlet delta oxygen (SDO). The latter is normally produced in a chemical SDO generator using very toxic and dangerous chemicals, which puts a limit for civilian applications of COIL that is still a very unique apparatus. Totally new non-chemical SDO generator is needed to allow oxygen-iodine laser to achieve its full potential as a non-hazardous efficient source of high-power laser radiation. There was interest in producing SDO in electric discharge plasma since the 50's long before COIL appearing. The idea of using SDO as a donor for iodine laser was formulated in the 70's. However, the injection of iodine molecules into a low- pressure self-sustained discharge did not result in iodine lasing. One of the main factors that could prevent from lasing in many experiments is a rather high threshold yield ~15% at 300K, which is needed for obtaining an inversion population. An analysis of different attempts of producing SDO in different kinds of electric discharge plasma has been done which demonstrates that high yield at gas pressure of practical interest (p > 10 Torr) for modern COIL technology can be obtained only in non-self sustained electric discharge plasma. The reason is that the value of relatively low reduced electrical field strength E/N ~10^{-16} V.cm^2, which is an order of magnitude less than that for the self-sustained discharge, is extremely important for the efficient SDO production. Although different kinds of non-self sustained discharges can be used for SDO production, we got started experiments with e-beam sustained discharge in gas mixtures containing oxygen. High specific input energy up to ~3 - 5 kJ/l. atm [O_2] has been experimentally obtained. Theoretical calculations have been done for different experimental conditions indicating a feasibility of reasonable SDO yield. Experimental and theoretical research of self-sustained electric discharge in SDO produced in a chemical generator, which is very important for getting plasma-chemical kinetic data needed for an estimation of SDO yield, is also discussed.

Keywords: COIL, iodine laser, singlet delta oxygen, electric discharge plasma

1. INTRODUCTION

Great success has been obtained in the research and development (R&D) of a chemical oxygen-iodine laser (COIL) [1] over the last 20 years, the best progress being obtained in the development of COIL for military applications such as the high-power airborne laser [2]. The potential of civilian applications of a COIL was claimed [3] to be very high because of its high-power output and the opportunity of using low loss optical fibers for delivery of the laser radiation at the lasing wavelength λ = 1.315 μm. Despite successful experiments on applications of COIL for construction materials cutting [4; 5], rock drilling [6], etc., COIL is still a very unique apparatus. The active medium of COIL is atomic iodine emitting photons when changing its electronic state: $I^*(^2P_{1/2}) \rightarrow I(^2P_{3/2}) + h\nu$. Atomic iodine in the excited state $I^*(^2P_{1/2})$ is produced by energy transfer from the singlet delta oxygen (SDO) $O_2(^1\Delta)$ molecule: $O_2(^1\Delta) + I(^2P_{3/2}) \rightarrow O_2(^3\Sigma) + I(^2P_{1/2})$ **(1).** SDO is obtained in COIL in a chemical generator through the following chemical reaction running in liquid phase: $Cl_2 + 2\,KOH + H_2O_2 \rightarrow O_2(^1\Delta) + 2\,KCl + 2\,H_2O$ **(2).** Very toxic and dangerous chemicals are used in the reaction, which put a limit for civilian applications of COIL. Thus, a totally new non-chemical SDO generator is needed to allow oxygen-iodine laser to achieve its full potential as a non-hazardous efficient source of high-power laser radiation. Quite a different method of SDO production can be used, namely, generation of SDO in electric discharge. The advantages of such a method of SDO production are the absence of dangerous chemicals, the feasibility of a development of a gas phase oxygen-iodine laser and other ones such as atomic iodine production through I_2 dissociation by $O_2(^1\Sigma)$ [7]. The problems of SDO production in electric discharge and development of an oxygen-iodine laser using SDO produced in electric discharge plasma, i.e. plasma-chemical oxygen-iodine laser (PlasmaCOIL), are discussed in the paper.

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2. SDO PRODUCTION IN ELECTRIC DISCHARGE

There was interest in producing SDO in electric discharge since the 50's long before the first laser was launched. "A fraction of $O_2(^1\Delta)$ between 5-25% of the total flow" was claimed to be observed in several papers [8] at gas pressure <1 Torr under RF or MW discharge excitation. For the first time the idea of using SDO as a donor for iodine laser was formulated in [9] a few years before the laser effect under chemical pumping, i.e. before COIL lasing was obtained. However, the injection of iodine molecules into a low- pressure self-sustained discharge (O_2:I_2; p≤1 Torr) did not result in iodine lasing. One of the main factors which could prevent from lasing in this and other experiments is a particular high threshold yield Y_{th}, which is needed for obtaining an inversion population $2[I(^2P_{1/2})]-[I(^2P_{3/2})]>0$, $Y_{th} = [O_2(^1\Delta)] / \{[O_2(^1\Delta)] + [O_2(^3\Sigma)]\} > (1 + 2\,K_{eq})^{-1}$, where $K_{eq} = [I(^2P_{1/2})]\,[O_2(^3\Sigma)] / \{[O_2(^1\Delta)]\,[I(^2P_{3/2})]\} = 0.75 \exp(\Delta E/T)$ is an equilibrium constant for the reaction (1); $\Delta E = 402$ K is the energy difference between $O_2(^1\Delta)$ and $I(^2P_{1/2})$ [10]. At T =300 K, $K_{eq} = 2.84$ and $Y_{th} = 0.15$; at T =100 K, $K_{eq} = 40$ and $Y_{th} = 0.012$. Another attempt of producing SDO by using non-self-sustained discharge [11] also failed, because the authors were unable to load any notable specific energy into the high pressure (1.18 bar) gas mixture of noble gas and electro-negative oxygen. DC self-sustained electric discharge was used for obtaining SDO in a set of papers (see [12,13], for instance), ~30% yield being claimed in the 80's [12] and ~10% at the end of the 90's [13]. No any attempt was made to get iodine lasing in these papers. There was quite recently demonstrated 21% yield under a MW discharge [14] and 32% yield under RF discharge excitation [15,16]. Iodine threshold lasing was claimed to be observed in the latter paper when mixing SDO with iodine, which looked more probably like iodine luminescence because there was no notable spectral line narrowing. However, the level of SDO partial pressure (< 1 Torr) in these [14-16] experiments seems to be of little any practical interest because the gas pressure should be ~ 10 Torr or higher for modern COIL technology. Moreover, for these experiments, estimation gives an energy efficiency for production of the SDO under conditions of maximum yield of about 2%. The reduced energy loading was about 50 J scm^{-3} (scm^{-3} means at unit gas volume at standard conditions, gas pressure 1 bar and temperature 300 K) that is 50 kJ/l atm at the same temperature. This is an enormous amount of energy almost completely released into useless gas heating.

Thus, up to now, there has not been any answer to the question, is there the possibility for efficient electrical excitation of SDO with a yield and partial pressure reasonable for development of a Plasma- Chemical Oxygen-Iodine Laser (PlasmaCOIL).

3. NON-SELF SUSTAINED DISCHARGE AS A POTENTIAL PRODUCER OF SDO

Quite recently it was claimed [7,17] that the value of reduced electrical field strength E/N (where E is electrical field strength, N is gas density) is extremely important for efficient SDO excitation. The parameter E/N must be ~ 10^{-16} V cm^2 (~2.7 kV/cm atm), which is an order of magnitude less than that for the self-sustained discharge used in most of the previous experiments.

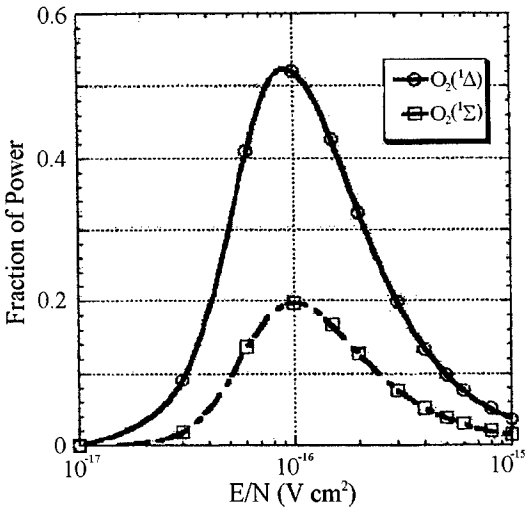

Fig.1. Fraction of power going into $O_2(^1\Delta)$ and $O_2(^1\Sigma)$ versus E/N [7]

This fact is illustrated by Fig.1 [7] that demonstrates a quasi-resonance dependence of fraction of electric discharge power going to production of SDO and $O_2(^1\Sigma)$. Therefore a non-self-sustained discharge that operates at low value of E/N parameter ~10^{-16} V cm^2 must be used for SDO production. A variety of non-self-sustained discharge can be used for such a purpose. A pulsed controlled avalanche discharge is used in Texas A&M University [17], SDO yield 16% being claimed to be obtained. A combined CW AC/RF discharge operating at low E/N parameter, in which RF electric field is applied to plasma generated by AC discharge, is used in the University of Illinois in Urbana-Champaign [7]. On the other hand, well-known e-beam sustained discharge (EBSD) lasers such as CO_2, CO and N_2O lasers, some of which use electronegative gases as an active medium, do operate at the same parameter E/N [18]. A feasibility of using EBSD for SDO production was discussed in [19-21]. Having very rich experience in R&D of such lasers and in study of EBSD in electronegative gases, our research team consisting of experimentalists of Gas Lasers Lab and Chemical Lasers Lab of the Lebedev Institute and theoreticians of TRINITI has started its activity to study both

experimentally and theoretically the best way of producing SDO with a high yield and at oxygen pressure adequate for development of Plasma COIL and to attempt to get PlasmaCOIL lasing.

4. EXPERIMENTAL AND THEORETICAL STUDY OF SDO PROPERTIES AND SDO PRODUCTION IN ELECTRIC DISCHARGE

To properly evaluate the concept of SDO production in EBSD it is necessary to perform experiments in conjunction with theory and modeling of the complex plasma discharge. Therefore, there should be performed following tasks: study of influence of SDO on electric discharge to get important kinetic data and development of EBSD generated plasma for oxygen-iodine laser.

4.1. Effects of SDO on electric discharge properties

Information about processes involving SDO and other excited states of oxygen molecule is rather scarce. A simple idea exploited by us is to place the self-sustained discharge chamber at the exit of traditional chemical SDO generator, which allowed us to vary the concentration of the SDO within a very wide range and study the influence of it on the discharge characteristics (Fig.2).

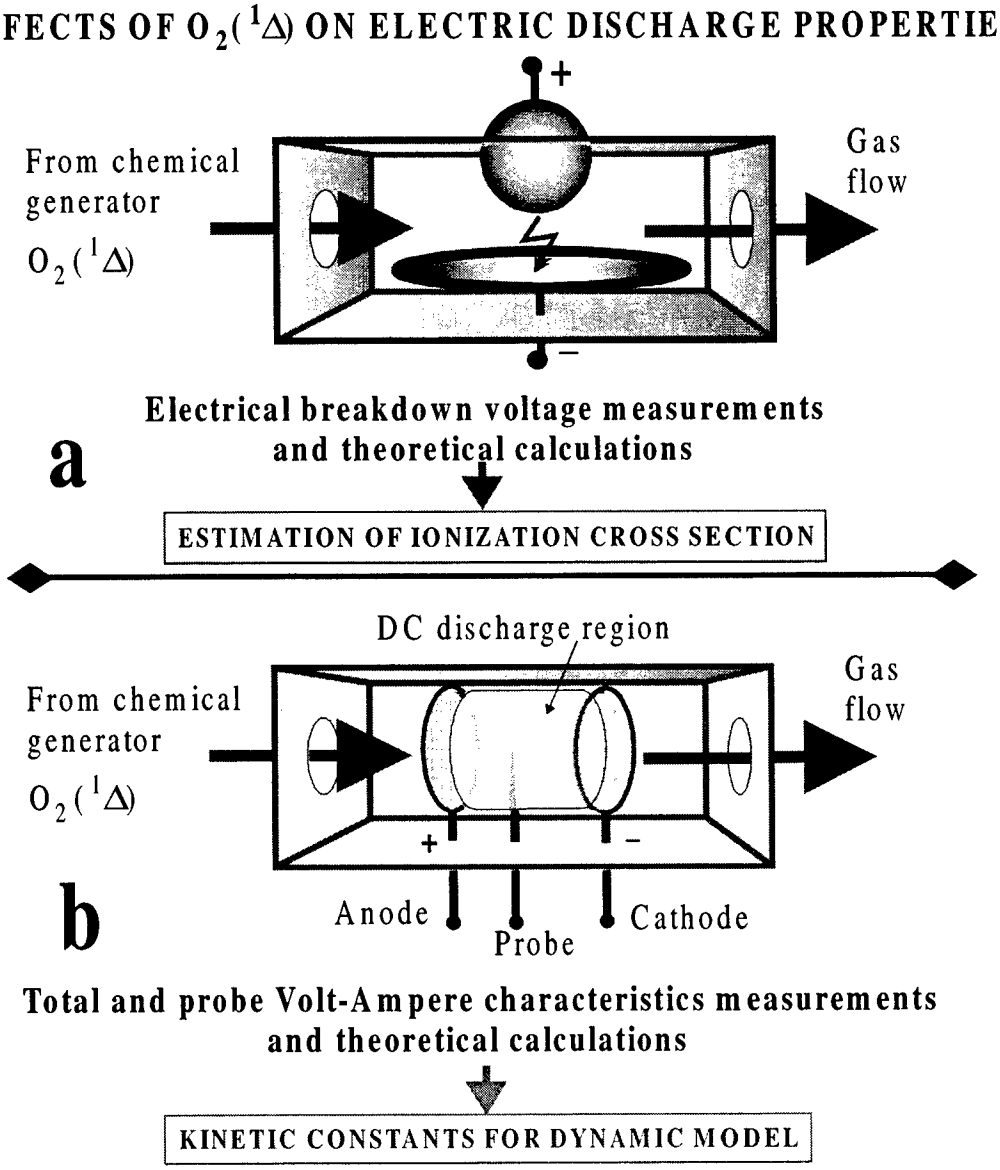

Fig.2. Experimental layout for electric discharge breakdown and volt-ampere characteristic measurements

4.1.1. Electric breakdown in SDO

Breakdown characteristics of a low-pressure self-sustained discharge in oxygen with 50% of SDO content were measured and compared with the breakdown characteristics of pure oxygen. The breakdown voltage as a function of the composition, temperature, and pressure of the gas mixture was measured in the discharge chamber connected to the gas duct downstream of SDO chemical generator. The electric field was directed perpendicularly to the gas flow (Fig.2a). The experimental results obtained were compared with the results of numerical modeling for adequate description of glow discharge in oxygen A comparison of breakdown electric field strengths for O_2 ($^3\Sigma$) and gas mixture $O_2(^3\Sigma)$:$O_2(^1\Delta)$ = 1:1 was done (Fig.3). The ionization cross-section appears to be of the most importance for calculations of electric breakdown parameters. The direct data on the ionization cross-section for SDO are absent. However, the position of the molecular terms for $O_2(^1\Delta)$ and $O_2(^3\Sigma)$ allows us to say that a simple downward shift along the energy axis gives satisfactory ionization cross-section for SDO.

$$\sigma_{ion}^{SDO}(E) = \sigma_{ion}^{O_2 ground}(E - \Delta E)$$

$$\Delta E = E_{SDO} - E_{O_2 ground} = 0.98eV$$

As one can see from Fig.3 threshold breakdown voltage

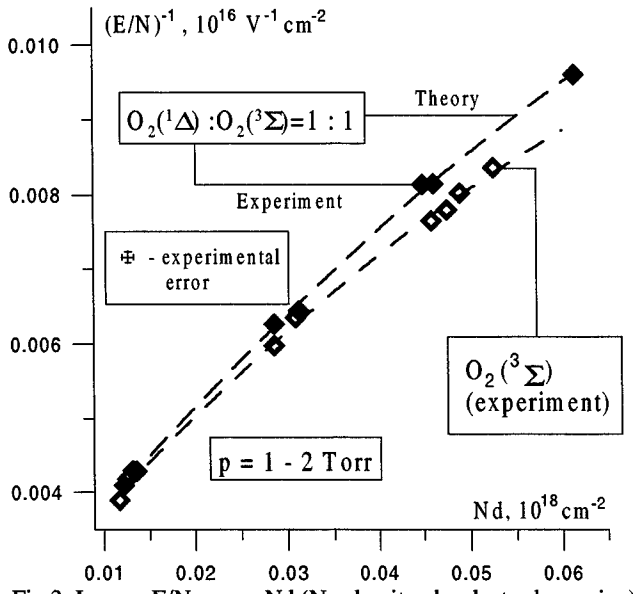

Fig.3. Inverse E/N versus Nd (N - density, d - electrode spacing) for electric discharge breakdown in oxygen and SDO (experiment and theory)

decreases when oxygen is excited on the singlet delta level. Good agreement between the experimental and theoretical data justifies the procedure of calculating σ_{ion}^{SDO}. A decrease in the breakdown voltage in the presence of $O_2(^1\Delta)$ is due to the increase in the ionization rate because of a lower ionization potential for the $O_2(^1\Delta)$ compared to $O_2(^3\Sigma)$.

4.1.2. Volt-ampere characteristics of electric discharge in SDO

The first results of experimental research of glow discharge in gas mixtures with high SDO content were obtained. Electric discharge chamber was incorporated into a gas duct going out of SDO chemical generator (Fig. 2b). An electric field being longitudinal relative to gas flow was produced with electrode system consisted of upstream anode and downstream cathode. The cathode was cooled by water. Electric potentials of plasma column in different points were measured with a metal needle probe. Experiments were carried out with pure oxygen going directly to the discharge chamber from a cylinder and gas mixture of oxygen containing 50% SDO going from SDO chemical generator through a water vapor trap to the discharge chamber. Probe and total volt-ampere characteristics of the electric discharge for pure oxygen and gas mixture O_2: SDO=1:1 at gas pressure of ~1 Torr

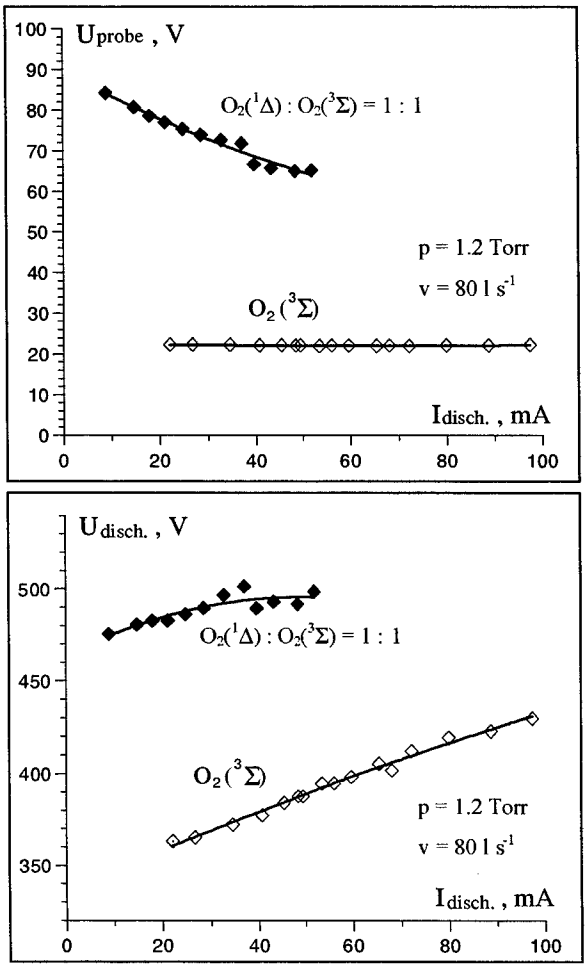

Fig.4. Volt-ampere probe and total characteristics of electric discharge in oxygen and SDO

are presented in Fig.4. As one can see in the figure, there is a big difference between the characteristics for pure oxygen and SDO, which indicates the fact that the two substances have different electrical features. It should be noted that the experimental results were obtained for relatively short electric discharge chamber, which did not permit to have a positive column of the electric discharge plasma. Although we did observe the big difference between the volt-ampere characteristics for pure oxygen and SDO, experiments should be done for longer discharge tube with a positive column, which makes it easy to calculate electric discharge characteristics. A simplified mathematical model for a description of a transitional space in electric discharge in oxygen was formulated. Diffusion coefficients, electron mobility and constants of ionization, attachment and detachment were calculated as a function of reduced electric field E/N. A comparison of the theoretical data with the experimental results is supposed to give new information about kinetic processes in electric discharge with SDO.

4.2. Electron beam sustained discharge in oxygen containing mixtures

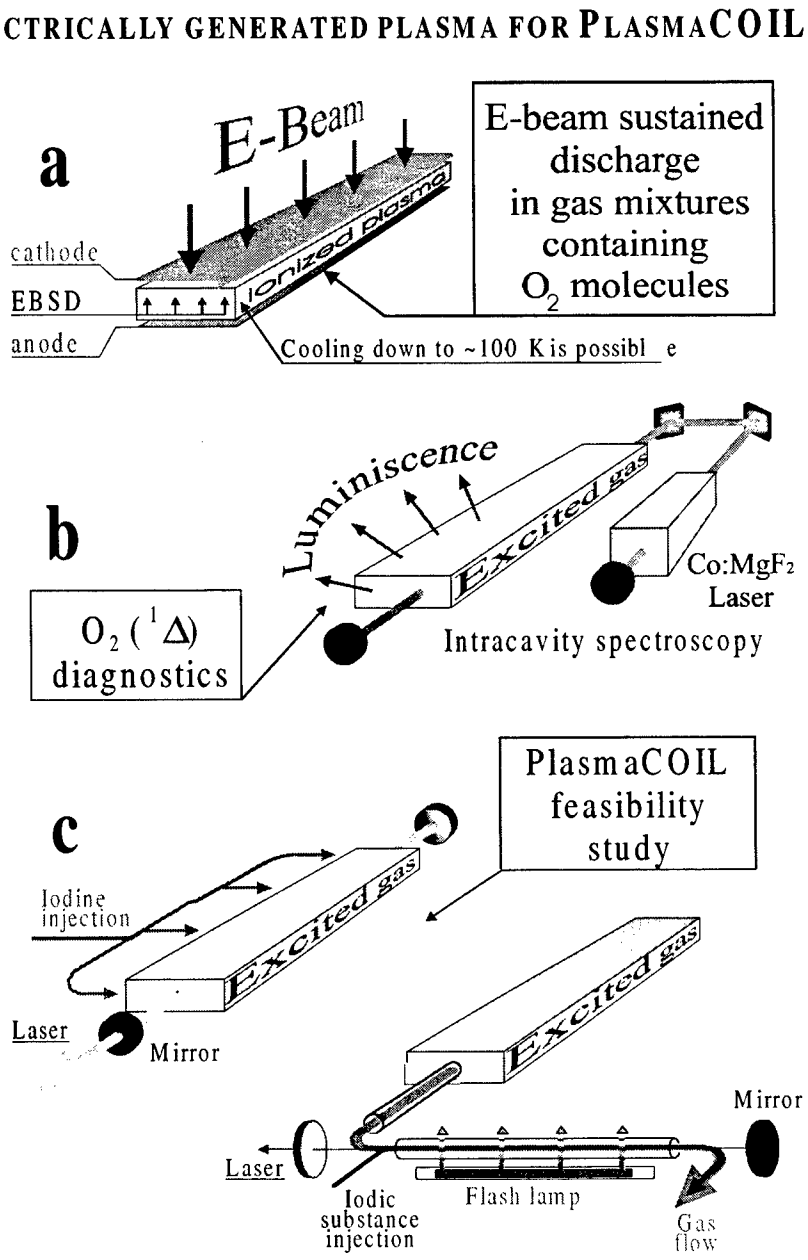

Fig.5. Schematic diagrams of EBSD application for SDO production and plasma-chemical oxygen-iodine lasing

One of the principal advantages of the EBSD approach (Fig.5a) is that it is possible to control E/N parameter within the E/N range most optimal for effective SDO production. However, there is an important question if it is possible at all to load much energy into electronegative oxygen in EBSD. Previously EBSD characteristics were studied both for electric discharge in pure oxygen [22] and its mixture with noble gases [11]. Taking into account a cathode voltage drop specific input energy into oxygen can be indirectly estimated as ~90 J/l atm [O_2] in [22] (active volume V=60 cm^3) and ~150 J/l atm [O_2] in [11] (V=150 cm^3; Ne:O_2=96:4; P=1.18 bar). It should be pointed out that minimal specific energy formally needed for transformation of all oxygen (with efficiency of 100%) from its ground state $O_2(^3\Sigma)$ into SDO state with energy of 0.98 eV is 94 kJ/mole [O_2] ≈ 4.2 kJ/l atm [O_2]. If one even assumes the efficiency of SDO production to be 100%, only 2-3% of oxygen is transformed into SDO in [11,22], which is an order of magnitude less than it is needed for oxygen-iodine laser. Quite recently we demonstrated experimentally [20,21] that far more specific energy up to ~3-5 kJ/l atm [O_2] can be loaded into EBSD with active volume of ~15 l (Fig.6). A fraction of total power going to SDO production taken from Fig.1 is also presented in Fig.6, which allows

one to estimate specific input energy going to SDO production ~1 kJ/l atm [O_2] corresponding to SDO yield up to ~25% . Of course, only experimental measurement of SDO concentration can give a final answer to the question if the EBSD is the best way of SDO production.

4.3. SDO diagnostics

The measurements of SDO concentration and gas temperature are the key elements of the measurement procedure. Different methods to measure the SDO concentration exist [8]. They are mass spectrometry, electron paramagnetic resonance, isothermal calorimetry, photoionization, "chemical" methods and optical spectroscopy [8]. Each of these techniques has limitations. For the pulsed discharge facility, the optical methods seem to be more preferable. Being strongly forbidden the $O_2(^1\Delta)\rightarrow O_2(^3\Sigma)$ transition emits very week radiation with the wavelength 1.27 μm. So, the very sensitive intrinsic germanium detectors cooled with liquid nitrogen or photon counting apparatus are needed to detect 1.27 μm radiation. These methods are applicable for a continuous process, but its application can be problematic when a pulsed process is under investigation. One of the problems is electric discharge interference. Nevertheless, it may be possible to use a liquid nitrogen cooled germanium detector with high sensitivity for measuring SDO concentration. However, intracavity laser spectroscopy (ILS) appears to be a very useful technique for measuring SDO and other

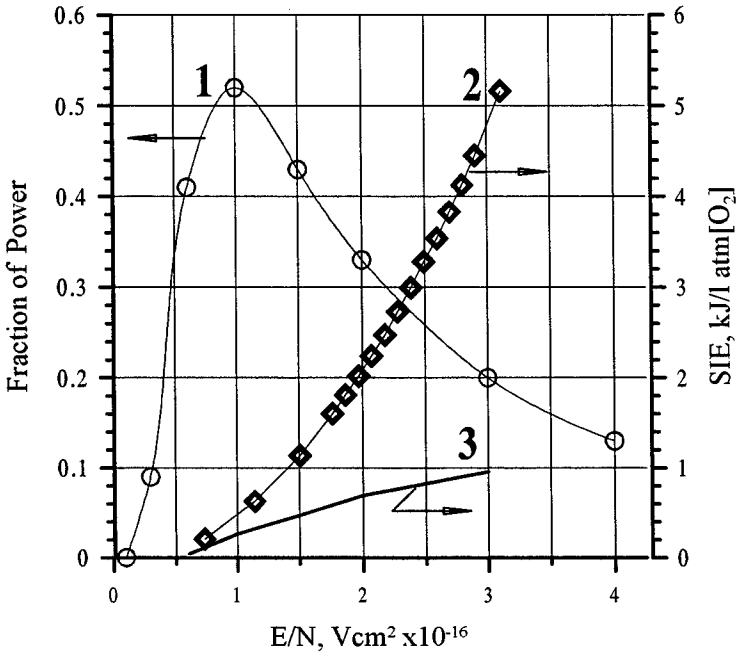

Fig.6. Fraction of power and specific energy going to oxygen and production of SDO. **1** - fraction of total power going to production of $O_2(^1\Delta)$[7]; **2** - specific input energy (SIE) going to O_2 (experimental data); **3** - SIE going to production of $O_2(^1\Delta)$

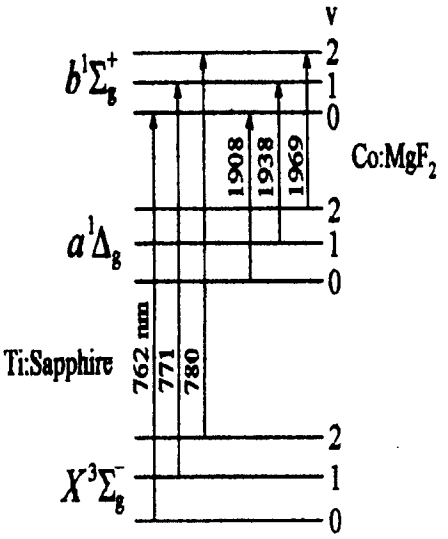

Fig.7. Schematic diagram of intracavity laser spectroscopy measurement of $O_2(^1\Delta)$ and $O_2(^1\Sigma)$ by Ti-sapphire and Co:MgF_2 lasers [23,24].

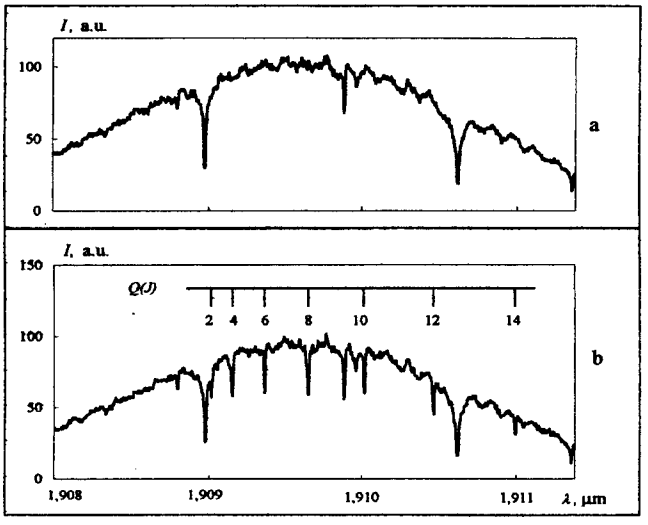

Fig.8. Output spectra of the broad band Co:MgF_2 laser (oxygen pressure in the laser cavity is 1.9 Torr): (a)- the discharge is turned off (absorption lines belong to water vapor); (b) - the discharge is turned on (additional absorption lines belong to the Q-branch of the 0-0 band of $a^1\Delta_g\rightarrow b^1\Sigma_g^+$ transition of molecular oxygen) [24].

species concentrations. The apparatus developed at the Lebedev Institute allows one to measure the concentration of the key components $O_2(^3\Sigma)$, $O_2(^1\Sigma)$, and $O_2(^1\Delta)$ [23,24] (Fig.5b). It was shown that ground state oxygen can be measured using a Ti: sapphire laser-based intracavity spectrometer operable near the 760 nm $O_2(^3\Sigma)\rightarrow O_2(^1\Sigma)$ absorption lines [23]. A sensitivity of ~(2-5) 10^{14} см$^{-3}$ was obtained when a Co:MgF_2 laser (1.6-2.5 μm tuning range) was used in the ILS method for direct measurement of $O_2(^1\Delta)$ produced in MW discharge [24]. The concentration of $O_2(^1\Delta, v=0,1,2)$ can be determined via absorption measurements at 1.91 μm (0,0), 1.94 μm (1,1) and 1.97 μm (2,2 bands of $O_2(^1\Delta)\rightarrow O_2(^1\Sigma)$ system) (Fig.7). Many rotational lines are recorded simultaneously in the ILS method due to the broad-band laser spectrum (Fig.8). Consequently, the gas temperature can be measured using the dependence of the absorption coefficients of molecular lines on the rotational quantum number.

Another approach is to inject iodine atoms to SDO and then detect the iodine luminescence at the wavelength 1.315 μm. As soon as iodine is in equilibrium with singlet oxygen due to the fast energy exchange process $O_2(^1\Delta) + I(^2P_{3/2}) \Leftrightarrow I(^2P_{1/2}) + O_2(^3\Sigma)$ the intensity of iodine luminescence can serve as a criteria of SDO yield (titration techniques). A calibration of this method can be made using a traditional chemical SDO generator with a known value of yield. The Einstein coefficient for iodine transition $I(^2P_{1/2}) \rightarrow I(^2P_{3/2})$ is more than four orders of magnitude larger than that of the $O_2(^1\Delta) \rightarrow O_2(^3\Sigma)$ transition. Thus, even at lower iodine concentrations of about 1% of that of oxygen the luminescence intensity can be three orders of magnitude higher. To avoid the problems of dissociating iodine molecules we suggest to mix oxygen extracted from the discharge region with iodide CF_3I, CH_3I and then decompose iodide molecules by photolysis. Photolysis is preferable to a discharge because of its high selective interaction with the active medium. As a final step of the experiments, atomic iodine can be mixed with the SDO produced in a pulsed EBSD plasma and small-signal gain and/or lasing if any can be measured (Fig5c).

4.4. Theoretical calculations of EBSD generated plasma

Electron energy balance as a function of the reduced electric field strength *E/N* is illustrated in Fig.9. It was calculated from solving the steady-state electron Boltzmann equation with the set of cross sections. Calculations were made for pure oxygen, all molecules in ground state. One can see that the energy fraction going into direct excitation of $O_2(^1\Delta_g)$ is fairly high approaching maximum 0.43 at $E/N = 0.87\ 10^{-16}$ V cm^2.

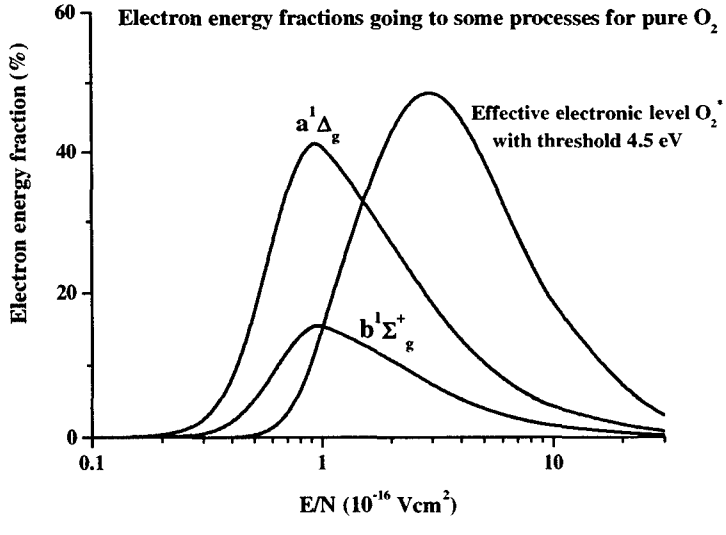

Fig.9. Electron energy fraction going to some processes for pure O_2

For the plasma with external source of ionization (e-beam, ionizing high-voltage pulses, etc.) the value of *E/N* can be controlled independently of electric current. Then, assuming that optimum conditions for production of the SDO are realized in a discharge plasma, we can take as an upper limit for SDO production efficiency, for a final concentration of 30%, the value η_{SDO}^{max}=0.21, which is roughly an average of initial and final (set equal to zero) efficiencies of direct excitation of $O_2(^1\Delta_g)$. The minimum energy loading for such an efficiency and SDO yield of 30% is easily evaluated: ε_{min}=6.5 J/scm^3. Assuming that the fraction of energy (1 - η_{SDO}) is released into gas heating, the increment of gas temperature can be estimated as 5500 K.

The above estimations allow us to immediately draw two conclusions: it is necessary to cool the gas flow; it is necessary to take special measures for discharge stabilization. Let us note that these evaluations may over-estimate gas heating (we neglect the energy contained in atoms and other excited states). From the other side, the total efficiency of SDO production may be higher

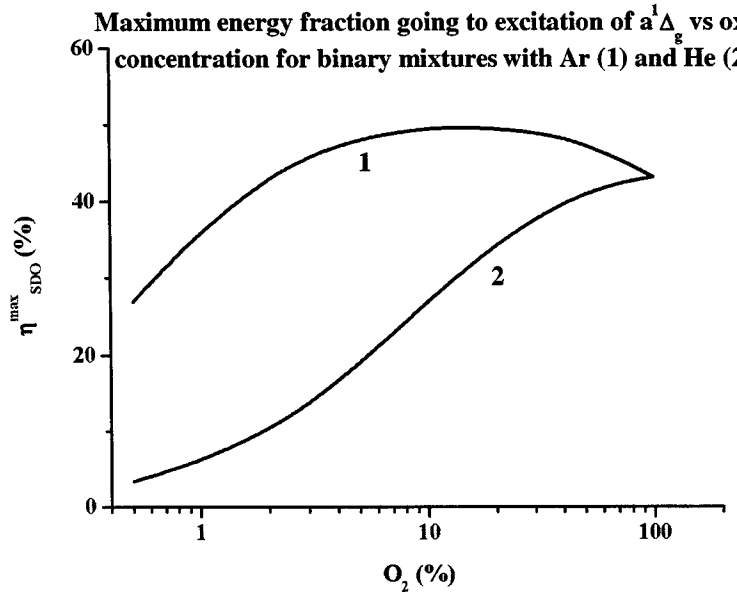

Fig.10. The maximum energy fraction going to excitation of SDO versus oxygen concentration for binary mixtures with Ar (1) and He (2)

than it was assumed (cascade excitation processes were neglected in making the estimations). Therefore, more detailed simulations may give more optimistic predictions.

Using mixtures of oxygen with some gases results in a variation of the electron energy balance. Generally, one may expect a decrease of the electron energy fraction going into direct excitation of $O_2(^1\Delta_g)$ when molecular or atomic additives appear. Let us note an interesting effect observed in gas mixture O_2:Ar. The results of Boltzmann calculations of η^{max}_{SDO} for this mixture in dependence on oxygen content are shown in Fig. 10. It is seen that an optimum mixture composition exists ($[O_2]\approx 0.1\ N$, where N is the total gas density and the brackets denote the oxygen molecule number density) for which $\eta^{max}_{SDO}=0.497$. For comparison, in Fig.10 a similar dependence of the maximum energy fraction going to excitation of $O_2(^1\Delta_g)$ on oxygen percentage in the binary mixture O_2:He is shown. For this mixture, η^{max}_{SDO} is a monotonously growing function of oxygen content. Use of the gas mixture O_2:Ar may be advantageous due to some increase in discharge efficiency and lowering of the gas temperature for the same energy load into singlet oxygen.

5. CONCLUSIONS

Breakdown and volt-ampere characteristics of self-sustained electric discharge in SDO studied both experimentally and theoretically indicate the fact that SDO and pure oxygen have different electric features. A comparison of the experimental and theoretical results justifies the procedure of calculating SDO ionization cross-section, the latter being that of ground state oxygen shifted 0.98 eV toward the lower energies. A difference between volt-ampere characteristics can give the opportunity of obtaining kinetic constants for SDO.

An analysis of different attempts of producing SDO in different kinds of electric discharge plasma has been done which demonstrates that high yield at gas pressure of practical interest (p > 10 Torr) for modern COIL technology can be obtained only in non-self sustained electric discharge plasma. The reason is that the value of relatively low reduced electrical field strength E/N ~10^{-16} V.cm^2, which is an order of magnitude less than that for the self-sustained discharge, is extremely important for the efficient SDO production.

High specific energy up to 3- 5 kJ /l atm[O_2] has been demonstrated to be loaded into a pulse non-self-sustained (e-beam sustained) discharge in oxygen containing mixtures, which together with theoretical calculations brings a hope of obtaining SDO yield in EBSD up to 20-30% with a high efficiency.

In case of positive results on mixing iodine with products of e-beam sustained discharge containing SDO, i.e. iodine lasing, well developed technology of EBSD CO_2 and CO lasers can be used in future for designing SDO generators for PlasmaCOIL. The latter laser technology being cryogenic can diminish demand in high SDO yield.

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